Photoinduced Electron Injection and Charge Separation in a Dye-sensitized Photoanode: A Quantum-classical Semiempirical description

Jan Paul Menzel[a] and Francesco Buda[a]

[a] *Leiden Institute of Chemistry, Universiteit Leiden, The Netherlands*
j.p.menzel@lic.leidenuniv.nl

Increasing energy demand, environmental concerns and rising CO_2 levels demonstrate the need to transition from a fossil to renewable fuel and energy economy. Direct conversion of solar energy to high energy molecular fuels would facilitate such a transition. Promising devices for the conversion of visible light directly into molecular fuel are Dye-Sensitized Photoelectrochemical Cells (DSPECs), which have attracted much interest in recent years [1]. To fulfil the potential of DSPECs in producing clean photosynthetic fuels, more effort is required to increase their chemical stability and efficiency. Since the efficiency of these devices is closely linked to the crucial process of photoinduced charge separation and charge recombination, molecular components and interfaces need to feature optimal energy level alignement. Computational studies can provide insight into these fundamental processes and suggest design principles. However, given the complexity of the system, a good compromise between accuracy and computational cost has to be found. Here, we use a combination of Density Functional based Tight Binding (DFTB) [2,3] and an extended Hückel approach [4] to model photoinduced charge separation and electron injection from organic dyes into a TiO_2 electrode.

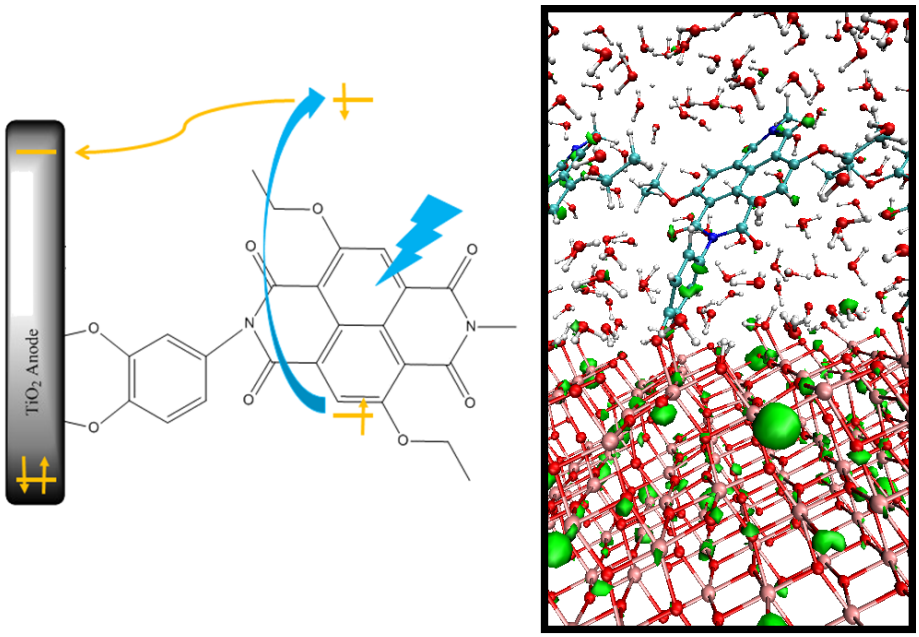

Figure 1: Electron injection from a photoexcited dye into a TiO_2-electrode

References

1. Z. Yu, F. Li, L. Sun, *Energy Environ. Sci.* **8** (2015), 760-775.
2. M. Elstner et al., *Phys. Rev. B* **58** (1998), 7260-7268.
3. AMS DFTB 2018, SCM, Theoretical Chemistry, Vrije Universiteit, Amsterdam, The Netherlands, http://www.scm.com
4. L.G.C. Rego, V.S. Batista, *J. Am. Chem. Soc.* **125** (2013), 7989-8997.